Experimental Results and Modeling

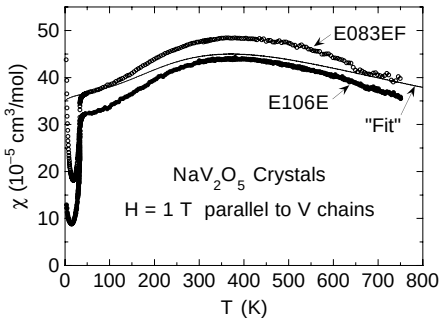
NaV_2O_5 Crystals
Magnetic Susceptibility

Modeling

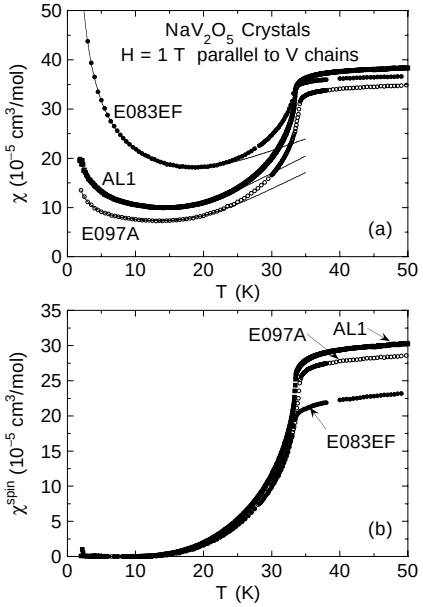
The observed susceptibility $\chi(T)$ is modeled using

$$\chi(T) = \chi_o + C_{imp}/(T - \theta_{imp}) + \chi^{spin}(T)$$

χ_o is the T-independent term
2nd term: Curie-Weiss impurity/defect term
3rd term: Theoretical spin susceptibility

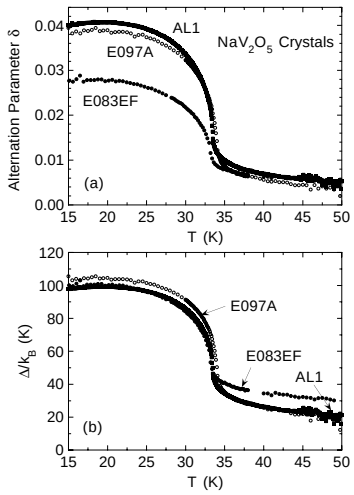


• The $\chi(T)$ of NaV_2O_5 crystals (symbols) above T_c = 34 K does not follow the prediction (solid curve "Fit") for the S = 1/2 AF uniform Heisenberg chain, contrary to claims in the literature
• In our modeling below, we assume that this is due to a temperature-dependent exchange constant above T_c



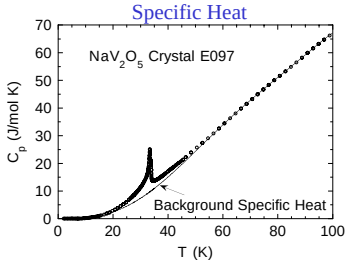
The $\chi(T)$ of three NaV_2O_5 crystals at low T is shown in (a). A spin gap develops below T_c = 34 K. The intrinsic spin susceptibility $\chi^{spin}(T)$ is obtained by subtracting the χ_o and Curie-Weiss impurity and/or defect contributions, as shown in (b)

The spin dimerization δ and spin gap Δ were extracted from $\chi^{spin}(T)$ using our theory:

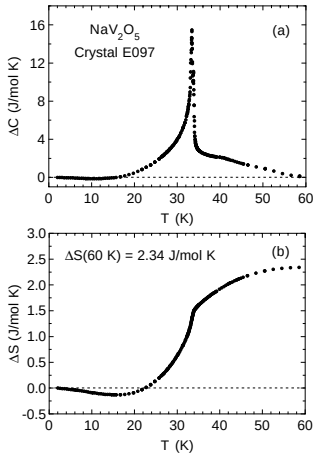


• The $\Delta(T = 0)$ = 100 K in (b) agrees with inelastic neutron scattering results. This is the first determination of $\Delta(T)$ directly from $\chi(T)$ data for any alternating-exchange chain
• A spin pseudogap of about 40 K is seen just above T_c, which decreases with increasing T. This is the first observation of spin dimerization order parameter fluctuations above T_c in NaV_2O_5

Specific Heat

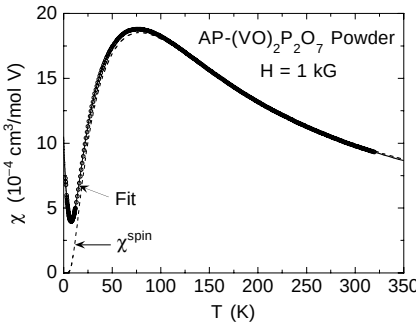


Changes in the specific heat ΔC and entropy ΔS due to the transition and associated order parameter fluctuations:



• The influence of spin, charge, and/or lattice order parameter fluctuations is seen in the lambda shape of the specific heat anomaly and ΔC from T_c up to at least 60 K
• At T_c, at least 77% of ΔS arises from lattice/charge degrees of freedom and less than 23% from spin degrees of freedom.
• Charge ordering and/or the lattice distortion strongly contribute to the thermodynamics of the transition

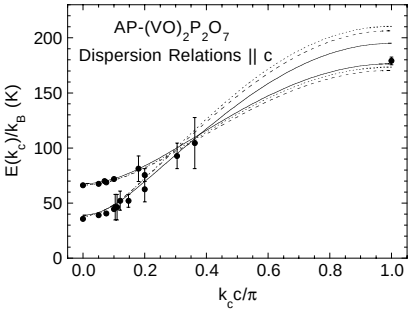
$\chi(T)$ of $(VO)_2P_2O_7$



Fit parameters for two distinct AF alternating-exchange chains:
Chain 1: J_1 = 130.3(7) K, J_2 = 109.5(9) K, Δ = 38.6(5) K
Chain 2: J_1 = 128.8(5) K, J_2 = 82.6(6) K, Δ = 67.5(5) K

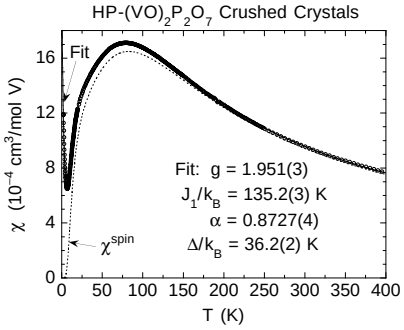
• The two-chain modeling would not have been possible without our high-precision theoretical work on the magnetic susceptibility of alternating-exchange chains
• The two-chain model is consistent with the $\chi(T)$ data
• The two spin gaps Δ are in agreement with neutron scattering data (see below)

One-magnon dispersion relations predicted for the two chains using the above exchange constants:



Neutron scattering data at 10 K (filled circles):
A.W. Garrett et al., PRL 79, 745 (1997)

• The agreement of our predictions (curves) with the data is good. Our prediction of two absorption peaks at k = π/c is in agreement with more recent neutron data (M. Enderle et al., unpublished)



• The fit in the figure is for a single type of alternating-exchange chain as demanded by the crystal symmetry
• A two-chain fit yielded the same parameters for each of the two chains as shown in the figure. This confirms the validity of our two-chain fit for $AP\text{-}(VO)_2P_2O_7$